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LASER PRODUCED X-RAYS FOR HIGH RESOLUTION LITHOGRAPHY. (U)  
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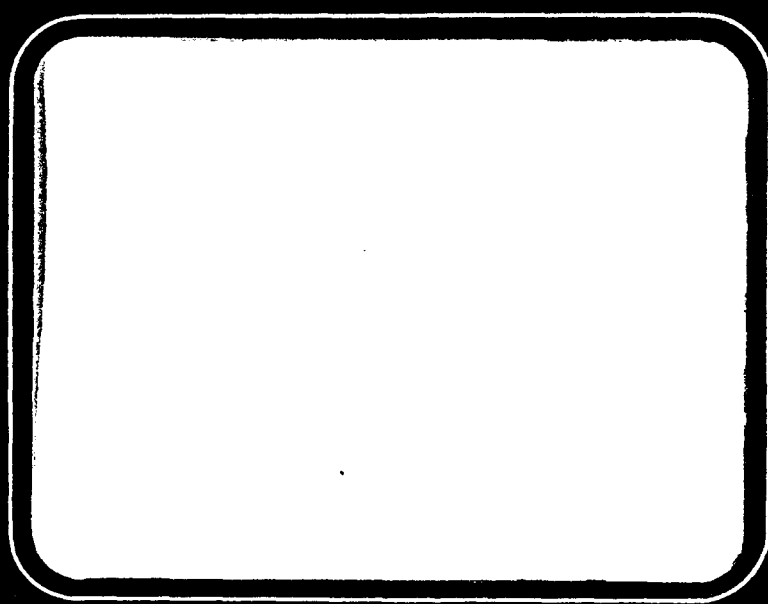
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20. higher average power should be available in the near future.



INTERIM REPORT

on

LASER PRODUCED X-RAYS FOR  
HIGH RESOLUTION LITHOGRAPHY

to

U.S. AIR FORCE  
OFFICE OF SCIENTIFIC RESEARCH

August 3, 1982

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Chief, Technical Information Division

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Dr. Mike Strocchio  
Air Force Office of Scientific Research  
Building 410  
Bolling Air Force Base, D.C. 20332

**Dear Mike:**

Battelle's Columbus Laboratories is pleased to submit for your consideration six (6) copies of the Interim Report of our research entitled "Laser Produced X-Rays for High Resolution Lithography".

This report reflects results achieved to date on AFOSR Grant No. AFOSR-82-0066. The key results are described in the section entitled "Results of Current Research".

Should you have any questions of a technical nature on this report, please contact Dr. Harold M. Epstein at (614) 424-5661. Questions of a contractual nature should be directed to Mr. D. H. Owens at (614) 424-5637. We are very encouraged by the early results of this program and are looking forward to continuing the research.

Sincerely,

Sincerely,  
Hal Epstein

Dr. Harold M. Epstein  
Senior Research Scientist  
Physical Sciences Section

HME:CS

Enclosure (6)

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### ABSTRACT

It has been demonstrated that a relatively small, high repetition rate laser can be a most attractive high average power source of x-rays in the 3/4 to 2 keV range. This x-ray energy range is particularly significant for x-ray microlithography of integrated circuits. Mode locked Nd-YAG lasers focused to several tens of  $\mu\text{m}$  spot sizes are very efficient x-ray sources for this purpose. Over ten percent of the laser light can be converted to x-rays of energy over 1 keV with a 400 mj 200 psec laser. Mode locked lasers with repetition rates of 10Hz and the above outputs are available now. Applicable laser systems with much higher average power should be available in the near future.

Conversion efficiency with 1.5 nsec pulses of 10 j have also been studied and preliminary results look very encouraging. However, the data analysis have not been completed, and more experimental work may be required. The high average power slab laser system under development at Stanford <sup>(1)</sup> is planned to produce 1.06  $\mu\text{m}$  pulses in this range.

INTERIM REPORT  
on  
LASER PRODUCED X-RAYS FOR  
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to  
U.S. AIR FORCE  
OFFICE OF SCIENTIFIC RESEARCH  
from  
BATTELLE  
Columbus Laboratories

August 3, 1982

INTRODUCTION

It is well established that x-ray lithography is an effective means for replicating sub-micrometer linewidth patterns<sup>(2)</sup>. Besides replicating test patterns, the technique has been used to fabricate surface acoustic wave devices, bubble domain devices, pn diodes, bipolar transistors, and MOS transistors. The basic concept of x-ray lithography is to use the short wavelength of an x-ray source instead of the long wavelength of an ultraviolet source. This essentially eliminates the diffraction limitation of the ultraviolet source. With this eliminated, x-ray lithography is capable of producing line patterns with a "line width accuracy" of less than 0.1  $\mu\text{m}$ .

The laser-plasma x-ray source has developed into the most intense laboratory x-ray source available in the energy range of  $\sim 3/4$  to 2 keV. In addition it appears to be the most attractive laboratory high-average-power x-ray source in this energy range, which is particularly significant for microlithography of integrated circuits. There can be little doubt about the need for high intensity and high-average-power x-ray sources which operate in this energy range, and which are small enough to be used in a laboratory or industrial locations. The rapid growth of synchrotron x-ray facilities demonstrates the increasing importance of high-average-power soft x-ray sources. However, the synchrotron is excluded as a labora-



tory x-ray source because of its size and cost. The commercial applications should expand greatly when the source becomes readily available at the home facility of the user.

The principal remaining problem in high resolution x-ray lithography is the development of an adequate soft x-ray source. The optimum x-ray energy for the few micron thick silicon masks and present photoresists is about 1.25 keV. The conventional rotating anode x-ray machines are very inefficient in this energy regime and have source diameters too large to produce high resolution at a reasonable distance. Although high conversion efficiency of laser pulses to x-rays was achieved more than 10 years ago, some of the research required to develop high-average-power sources in the most useful pulse energy ranges still remains to be done.

In this report, the development of the laser plasma x-ray source for soft x-ray lithography on integrated circuits will be discussed. This application utilizes the high-average-power of soft x-rays attainable with a rapidly pulsed laser of smaller pulse size. Other applications such as microradiography of thin samples, EXAFS (extended x-ray absorption fine structure), and medical treatment should also benefit indirectly.

#### BACKGROUND

This section discusses the basis of plasma x-ray sources with emphasis on the laser-plasma source<sup>(3-9)</sup> as applied to x-ray lithography applications.

### Comparison of Plasma Sources

Comparison of various plasma x-ray sources for lithography application is complex because at least three plasma parameters are involved: plasma temperature, radiating area, and conversion efficiency of energy absorbed in the target to plasma x-rays. Almost all plasma x-ray sources of interest for x-ray lithography are thin radiators in the energy range of interest. That is, the reabsorption of the x-rays by the plasma is negligible, and the dominant spectral shape of the emitted x-rays follows the plasma bremsstrahlung envelope and not that of a black body emitter. This does not mean that K, L, or M lines will be absent from the plasma. They will be present in large numbers but the statistical envelope of the radiation energy intensity will have the approximate shape  $\exp(-h\nu/kT)$  where  $h\nu$  is the photon energy in keV and  $kT$  is the plasma thermal energy in keV.

The energy  $E_p$  incident on the photoresist is given by

$$E_p \approx C_1 / 2\pi r^2 \int_0^\infty \exp[-(\alpha x - (h\nu)/kT)] d(h\nu) \text{ J/cm}^2 \quad (1)$$

where  $\alpha(\text{cm}^{-1})$  and  $x(\text{cm})$  are the absorption coefficient and thickness respectively of the open part of the mask,  $r$  is the distance from source to photoresist and  $C_1$  is a constant depending on the laser pulse energy, pulse width, and target. For laser pulses on copper, which are currently being considered for the high average power source,  $C_1$  is approximately  $.1E_L$ . For a silicon mask below the K-edge,  $\alpha \approx 5000(h\nu)^{-3}$ . With these assumptions we find,

$$E_p \approx .1 E_L / (2\pi r^2) \int_0^\infty \exp[-5000(h\nu)^{-3} x - (h\nu)/kT] d(h\nu) \text{ J/cm}^2 \quad (2)$$

The x-rays emitted from the plasma fall off exponentially with increasing energy, while the x-ray transmissivity of the mask,  $T(h\nu)$  is controlled by the photoelectric absorption cross-section and falls off rapidly with decreasing energy. The resultant x-rays reaching the photoresist fall in a narrow band as shown in Figure 8, and for the purposes of this analysis can be treated as monoenergetic with energy given by the peak  $(h\nu)_m$ . For silicon this is given by

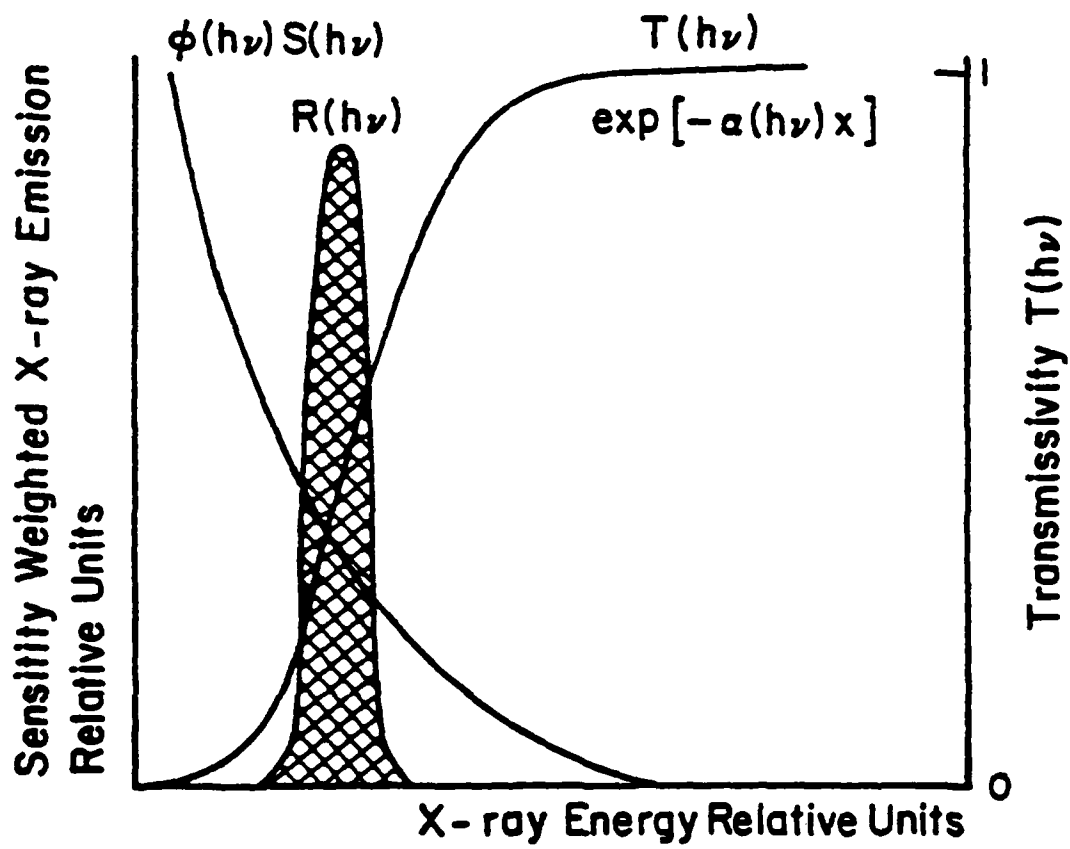


FIGURE 1. SENSITIVITY WEIGHTED X-RAYS ON PHOTORESIST

$$(h\nu)_m = 11 \times 1/4 kT^{1/4} \quad (3)$$

Because of the sharply peaked integrand, when  $kT$  is less than  $(h\nu)_m$ , Eq. 2 can be evaluated by expanding the integral in a Taylor series about  $(h\nu)_m$  (saddle-point method), giving

$$E_p \approx .07 E_L r^{-2} (kT)^{5/8} x^{1/8} \exp(-14.7 x^{1/4} / (kT)^{3/4}) J/cm^2 \quad (4)$$

For a 3 micrometer silicon mask and an 0.9 keV temperature plasma ( $t_p$  is the duration of the laser-plasma),  $(h\nu)_m = 1.41$  keV. The peak is sufficiently below the 1.84 keV K-edge of silicon to permit our analysis to be valid. The amount of photoresist exposure which would have resulted from x-rays above 1.41 keV is small.

Now if we consider a typical photoresist, without absorption edges in the region of  $(h\nu)_m$ , the absorbed dose,  $D$ , is approximately proportional to  $(h\nu)^{-3}$ . Thus from Eqs. 3 and 4,

$$D \propto (kT)^{-1/8} x^{-5/8} \exp(-14.7 x^{1/4} / (kT)^{3/4}) \quad (5)$$

Since the coefficient of the exponent is only very weakly dependent on  $(kT)$ , the dose to the photoresist is essentially exponentially dependent on  $(kT)^{-3/4}$ . For a mask of 3 micrometers silicon and a  $(kT)$  of 0.9 keV, the exponential attenuation is 8.1. The silicon K-edge will cause severe attenuation if  $(kT)$  is increased by more than a factor of 2 and a  $(kT)$  of 1.8 keV still has an exponential attenuation of  $\sim 3.5$ . Therefore the dose for the present laser-plasma source is only a little more than a factor of 2 less than that of the optimum plasma bremsstrahlung spectrum. It is nearly an optimum source for lithography.

I know of no electrical spark source producible with machines of reasonable size and cost that will produce a plasma of more than about 0.1 keV. The x-rays emitted by this type of plasma will have an exponential attenuation of over  $10^4$ . Many discharge plasmas emit higher energy

non-thermal x-rays. But the spectrum normally includes a hard component which causes an unacceptably weak contrast between exposed and unexposed areas.

Two other factors must also be considered in comparing plasma sources: source size, and conversion efficiency of energy absorbed in the target to x-rays. The obvious effect of source size is on resolution. At a source-to-wafer distance,  $D$ , and a mask-to-wafer distance,  $S$ , a flat disc x-ray source of radius,  $r$ , produces a "blur",  $\delta$  of  $\delta = S(2r/D)$ . Probably of greater significance than the blur is the opportunity to bring the x-rays from a small source out of the vacuum through differentially pumped orifices and avoid the rather large x-ray attenuation through thick windows. Laser plasma sources are generally much smaller in volume than sparks.

The significance of the conversion efficiency of energy absorbed in the target to x-rays in a useful energy band is somewhat more subtle. A low conversion efficiency means that a large amount of target debris is generated with the x-rays. This debris can damage and plate windows in addition to damaging orifices for differential pumping. Even if a spark source produced sufficient x-rays for lithography, the debris would make it a difficult source to use.

#### Scaling Criteria

For the high-average power applications, it is advantageous to use a low pulse energy, high repetition rate laser. The high conversion rates of laser energy to x-rays (up to 27% over 300 eV) at Battelle have been achieved in the near steady-state coronal plasma radiation regime with carefully pre-conditioned plasma profiles. The rule of thumb dividing line between steady state and time dependent coronal plasmas is given by  $t \approx 10^{12}/n_e$ . Since the critical plasma density, where the principal absorption occurs is  $n_e \approx 10^{21}$ , radiation times greater than 1 nanosec can be reasonably assumed to be steady state. Actually a large fraction of the absorbed energy is conducted to, and radiated from, regions of greater density than  $10^{21}$ , so somewhat shorter irradiation times can still be considered likely to be described by a steady state

radiation model. This is an important factor if we are to assume, for scaling purposes, that the radiated x-ray spectrum depends only on the power density and electron density gradient at the critical-density surface. If quasi-steady state conditions have not been achieved, the spectrum will depend on previous irradiation history.

The focal diameter,  $D_f$ , at the critical surface of the plasma changes as the plasma moves out at a velocity  $v$ . If the laser is focused on the critical density surface at time  $t$ ,  $D_f$  is given by

$$D_f = f D_B \Delta + v|t|f \quad (1)$$

where  $f$  is the  $f$  number of a perfect lens,  $D_B$  is the incoming beam diameter,  $\Delta$  is the natural beam divergence, and  $v$  is the plasma velocity. The first term represents the initial focal diameter on the critical surface and the second is the increase in diameter due to the expanding plasma. The situation in which the laser is focused at the position which the critical density surface will occupy when the laser reaches peak power corresponds to  $t = 0$  when the laser power is at its maximum. However, for purposes of scaling, this is not a factor.

The power density at focus,  $P$ , is given by

$$P = 4/\pi [f^2 D_B^2 \Delta^2 / P_L + 2v|t|D_B \Delta / P_L + v^2 t^2 / (f^2 P_L)]^{-1} \quad (2)$$

where  $P_L$  is the laser pulse power. In the steady state  $v$ , is a function only of  $P$  for a given target. If  $f$  and  $\Delta$  are kept constant and  $t$  and  $D_B$  are scaled as  $P_L^{1/2}$ ,  $P$  will remain unaffected. Thus, the spectrum and conversion efficiency will be conserved with this scaling. This type of scaling has been tested at Battelle over a laser pulse energy range of over 100, and found to be valid.

For typical conditions of  $P \approx 2 \times 10^{13}$ , the copper plasma will be about 19 times ionized at the maximum plasma temperature of  $\sim 0.9$  keV and  $v \approx 3 \times 10^7$  cm/sec. The optimum  $f$  number for the lens is given by

$$f_{\text{opt}} = \left( \frac{vt}{D_B \Delta} \right)^{1/2} \quad (3)$$

Of course the f number of the lens cannot change with time on a nano-second time scale. For a high average power system we would like to have P exceed a threshold for an extended time rather than have the peak power coincide with the minimum focal diameter. This also keeps v relatively constant through a large part of the pulse. Plasma velocity is a weak function of P anyway. Choosing the lens to optimize the power at time  $\tau$  gives

$$P \approx \frac{P_L \tau}{\pi v D_B \Delta} \quad (4)$$

In scaling,  $\tau$  is proportional to the pulsewidth.

If high conversion efficiency is desired for low energy pulses, a short pulse width is required. However, it is not obvious why low energy pulses are desirable for a high average power laser source. Large pulse, solid state lasers are almost universally made of glass. Because of the thermal expansion and low pumping efficiency of glass, it is several times more expensive to obtain the same average power from a glass system than a YAG system. Unfortunately YAG rods are limited to small diameters (1 cm or less for commercial systems). It is, therefore, advantageous to be able to operate with low energy pulses. Since our past experience has shown that P should exceed  $\sim 2 \times 10^{13} \text{ W/cm}^2$ , a pulse energy under 1 Joule requires a pulse width less than 1 nanosec. Such a pulse width can only be attained on commercial systems by mode locking, and available mode-locked systems become unstable at pulse widths longer than  $\sim 0.25$  nanosec. At this pulse width the  $\sim 1$  cm diameter YAG rod is limited to 0.3-0.4 Joules per pulse if long life is required. These conditions are more than adequate to exceed the requirement that  $P \gtrsim 2 \times 10^{13}$  but conversion efficiencies of laser light to x-rays of the desired energy have not been determined for pulses of this low energy in the time dependent coronal regime prior to the present study.

### RESULTS OF CURRENT RESEARCH

The results of the research to date in this program have been very encouraging. It was shown in the discussion on laser energy scaling that the desired conversion efficiencies should be attainable if the following scaling condition is met.

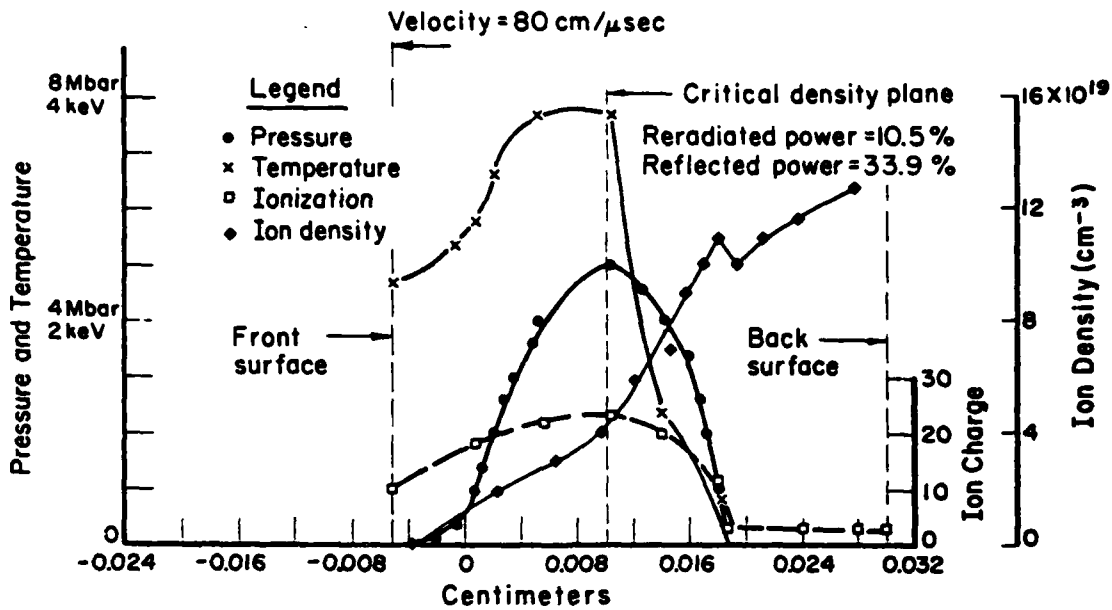
$$E_L / v \tau^2 D_B \Delta \gtrsim 10^{14}$$

where  $E_L$  is the laser energy in joules,  $\tau$  is the width of the laser pulse at half maximum,  $v$  is the plasma velocity in cm/sec,  $D_B$  is the beam diameter in cm, and  $\Delta$  is the beam divergence in radians. This condition assumes an optimum lens and plasma steady state conditions, as well as optimum preparation of the plasma at the surface of the target. Since a high repetition rate mode locked laser is now available commercially with an energy of 400 mJ at a pulse width of 0.2 n sec and a pulse rate of 10 Hz, these single pulse conditions were chosen for our experimental study. To go from single pulse x-ray production to a repetitive pulse system it is only necessary to devise a rapid target changing mechanism.

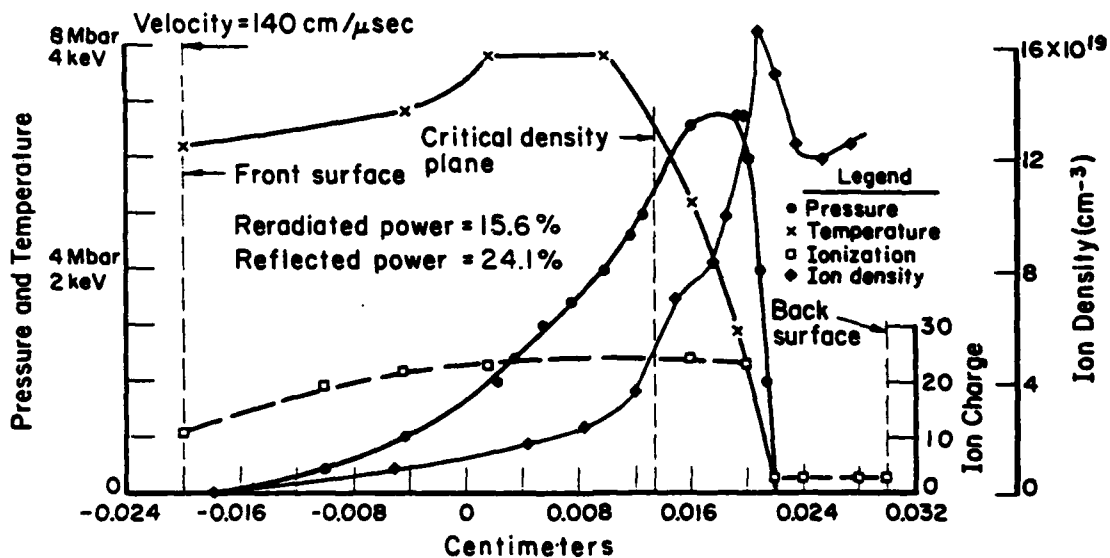
The mode locked laser pulse conditions easily meet the scaling condition equation. The major unknown factors are whether the plasma approaches steady state conditions in 0.2 n sec and whether the correct surface plasma can be realistically produced. A computer analysis done with Battelle's Flash code shows the state of ionization as a function of time for a 1-dimensional constant laser irradiation on iron assuming time dependent state ionization conditions. It can be seen from Figure 2 that the time dependent ionization has reached over 90 percent of its nanosecond value in 265 picoseconds. Since the ionization levels involved are closely spaced L levels, one would not expect to see a significant decrease in the energy of the line radiation.

The experimental results showed this conclusion to be valid in the range of interest. As shown in Table I, the conversion efficiency of laser energy to x-rays up to 2 keV is about the same for several hundred mJ, 200 picosecond pulses as for 100 J, 1.5 n second pulses.



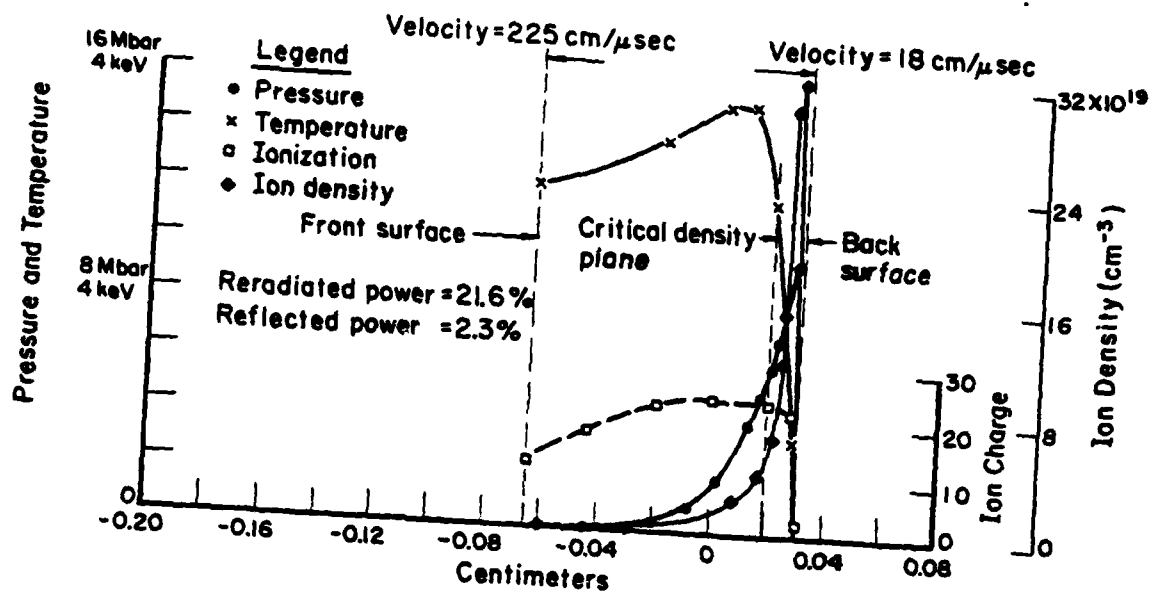


(c) Time = 135 Picoseconds

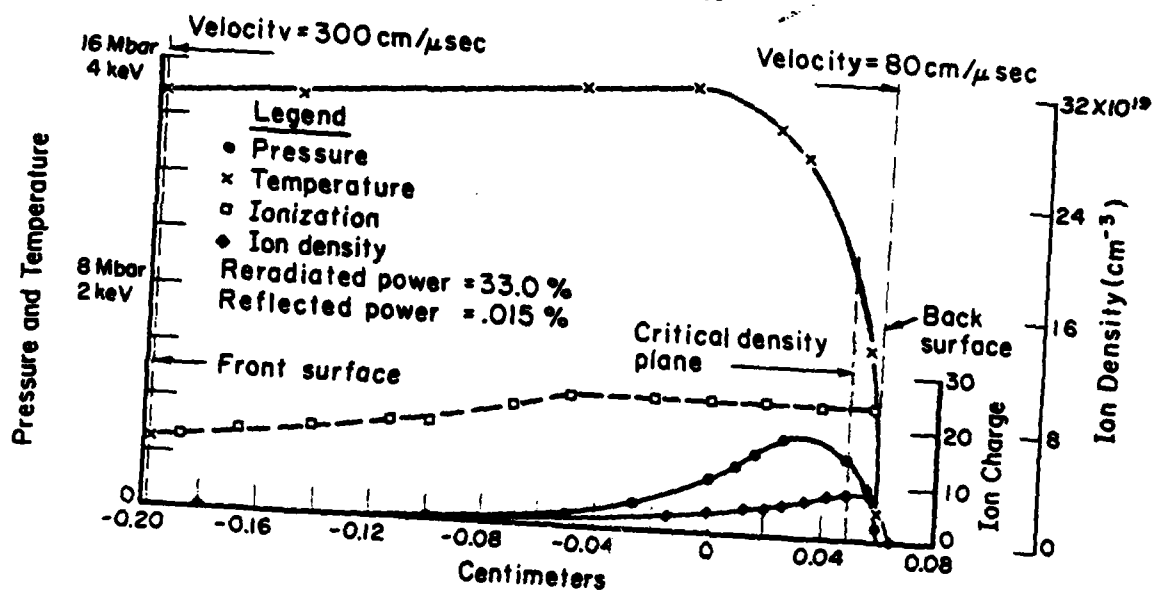


(d) Time = 265 Picoseconds

FIGURE 2. Time Dependent Coronal Model



(e) Time = 500 Picoseconds



(f) Time = 1000 Picoseconds

FIGURE 2. (CONTINUED)

TABLE I. Comparison of Recent 500 mj Subnanosecond  
with Previous 1.5 Nanosecond 100 j Tests

Be-Thickness, mils	Effective Energy keV	Conversion Efficiency Above Effective Energy	
		500 mj-200 psec	100 j-1.5 nsec
5	1.21	0.12	0.096
1	1.43	0.050	0.058
2	1.71	0.032	0.032
3	1.89	0.020	0.021
5	2.14	0.008	0.012
10	2.55	0.004	0.0053
15	2.82	$1 \times 10^{-4}$	0.003

TABLE II. Comparison of Recent 200 mj Subnanosecond  
Differential Foil Spectra with Previous  
1.5 Nanosecond, 100 j Tests

Be Thickness Mils	Effective Energy keV	Conversion Efficiency Above Effective Energy	
		.26 j-200 psec	100 j-1.5 nsec
1/2	1.21	.12	.096
1	1.43	.049	.058
2	1.71	.031	.032
3	1.89	.0065	.021
5	2.14	$7.7 \times 10^{-4}$	.012
10	2.55	$2.5 \times 10^{-4}$	.0053
15	2.82	$2.4 \times 10^{-5}$	.003

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#### FUTURE WORK

The key objective of this program, obtaining high conversion efficiencies with laser pulses of less than one joule, has already been attained. The remaining research program deals primarily with the use of multiple element targets to improve the conversion efficiency of x-rays into the 1-1.5 keV energy band. The target to be used will be available or easily cast alloys of which could include elements such as Cu, Fe, Ni, Zn, Mn, Cr, Al, and Mg. All of these have neither L or K lines in the energy regime of interest.

The results of this program have been very encouraging and a journal article on the program will be submitted for publication this year.